Supplementary Information

Ultrafast Self-Powered Semi-Transparent Pyro-Phototronic Device Array Based on Room-Temperature Amorphous In_2S_3/NiO Heterostructures†

Malkeshkumar Patel,[a,b,ψ] Shubham Umeshkumar Gupta,[a,b,ψ] Vivek Mohan More,[a,b] Sourov Hussain,[a,b] Seojun Jeon,[a,b] Kyu-Won Kim,*[c] and Joondong Kim,**[a,b]

[a.] Photoelectric and Energy Device Application Lab (PEDAL), Multidisciplinary Core Institute for Future Energies (MCIFE), Incheon National University, 119 Academy Rd. Yeonsu, Incheon, 22012, Korea (Rep. of).

[b.] Department of Electrical Engineering, Incheon National University, 119 Academy Rd. Yeonsu, Incheon, 22012, Korea (Rep. of).

[c.] Department of Chemistry, Incheon National University, 119 Academy Rd. Yeonsu, Incheon, 22012, Korea (Rep. of).

Email: J. Kim (joonkim@inu.ac.kr) and (K. Kim) kyuwon_kim@inu.ac.kr

ψ There authors contributed equally.

Device Fabrication: Glass coated with fluorine-doped tin oxide (FTO) (sheet resistance ≈ 7 Ω/sq) and Eagle glass were used as substrates. Prior to deposition, substrates were cleaned sequentially in acetone, methanol, and deionized water, each step lasting 10 minutes in an ultrasonic bath, then dried with a nitrogen stream. The amorphous In_2S_3 layer was deposited by magnetron sputtering using an In_2S_3 target (diameter ∅2 inch, iTASCO, purity 99.99%). Sputtering conditions were RF power 50 W, chamber pressure 5 mTorr, and argon flow 30 sccm. The In_2S_3/NiO heterojunction was built layer-by-layer by sputtering NiO onto the In_2S_3 layer. The In_2S_3 synthesis parameters were kept identical to those described above, yielding a growth rate of about 1.5 nm min^{-1} for In_2S_3. NiO was deposited by reactive sputtering from a Ni target in an oxygen-containing ambient. Sputtering power: 50 W (DC); gas composition: Ar 20 sccm, O_2 5 sccm; total pressure ≈ 3 mTorr; growth rate ≈ 2 nm min^{-1}. Pre-sputter cleaning was 3 minutes, and the substrate holder was rotated at 5 rpm to promote uniform coverage. A network of silver nanowires (AgNWs, ~25 nm diameter, ~25 μm length) was prepared in isopropanol and spun onto the substrate at 2500 rpm for 30 seconds to form a percolating film with tunable density by solvent adjustment. A laser-patterned PET mask defined the top electrode region. The AgNW network was thermally treated at 100 °C to remove residual solvent and promote networking. A ZnO protective layer (10–15 nm) was then deposited atop the AgNW network to complete the semi-transparent electrode stack. ZnO was sputtered from a 99.99% purity ZnO target under these conditions: RF power 30 W, Ar flow 50 sccm, pressure 5 mTorr, duration 10 minutes. The sputtering base pressure remained at 2.5×10^{-6} Torr, and the substrate was rotated at 5 rpm during deposition.

Characterizations: The crystallinity and phase of the In_2S_3 films were probed by X-ray diffraction using Cu Kα radiation (λ = 1.5405 Å) in grazing incidence mode at a glancing angle of 1°. The XRD setup employed 45 kV, 200 mA, with an incident slit 1 mm wide; 2θ steps of 0.02° were collected, and a moderate scanning speed was used. Transmittance, reflectance and absorbance spectra were recorded

with a UV-Vis-NIR spectrophotometer (Shimadzu 2600 UV) equipped with a diffuse reflectance integration sphere. Baseline correction was performed using a blank in air, and measurements spanned 250–1400 nm. Surface morphology was examined by field-emission SEM (JSM-7001F, JEOL). Elemental composition and distribution were assessed by energy-dispersive X-ray spectroscopy (EDS) to confirm In and S presence and uniformity. For electrical and optoelectronic measurements, current density–voltage (J–V) characteristics were obtained with a potentiostat/galvanostat (PGStat, WonA Tech). Illumination intensity was controlled via a dual adjustable power supply and quantified with calibrated power meters and a Si photodiode. Transient response and timing were evaluated under pulsed illumination using chronoamperometry, driven by a function generator (Topward 8150) with a 50% duty cycle square wave. Transient photocurrent measurements were conducted using 420 nm LED sources. A wavelength-tunable LED source system (WLS-22-A, Mightex) and a PV power meter (K101, McScience) were used to benchmark device performance.

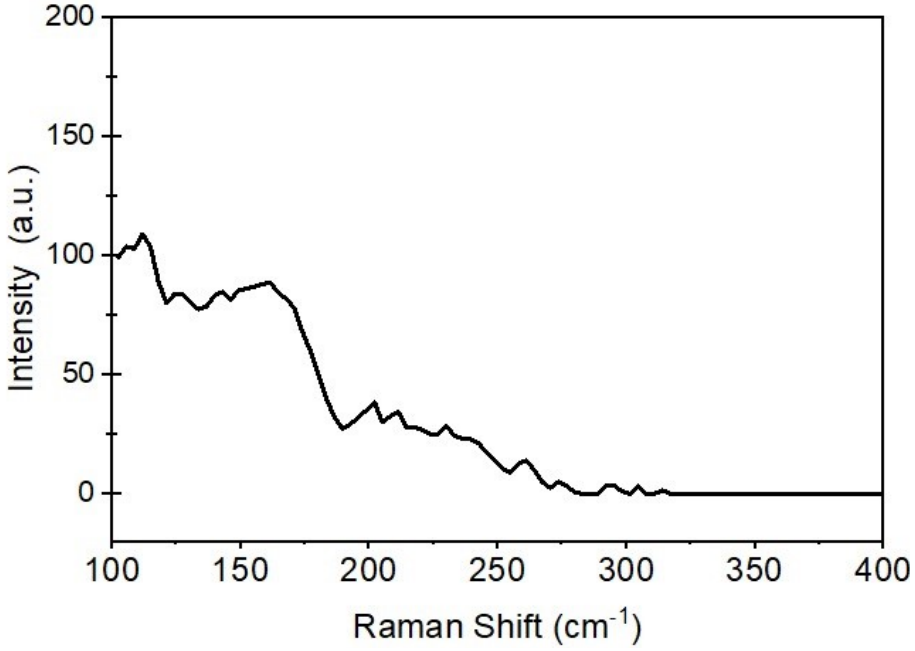

Figure S1. Raman spectra of In_2S_3 film.

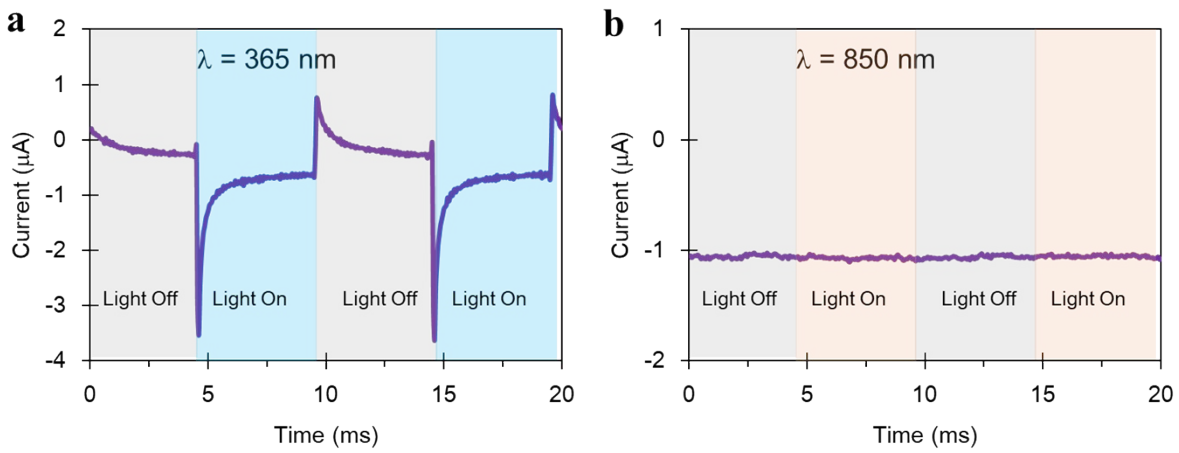

Figure S2. Spectral photoresponse of In_2S_3/NiO device under the pulsed light illumination wavelength of (a) 365 nm, and (b) 850 nm.

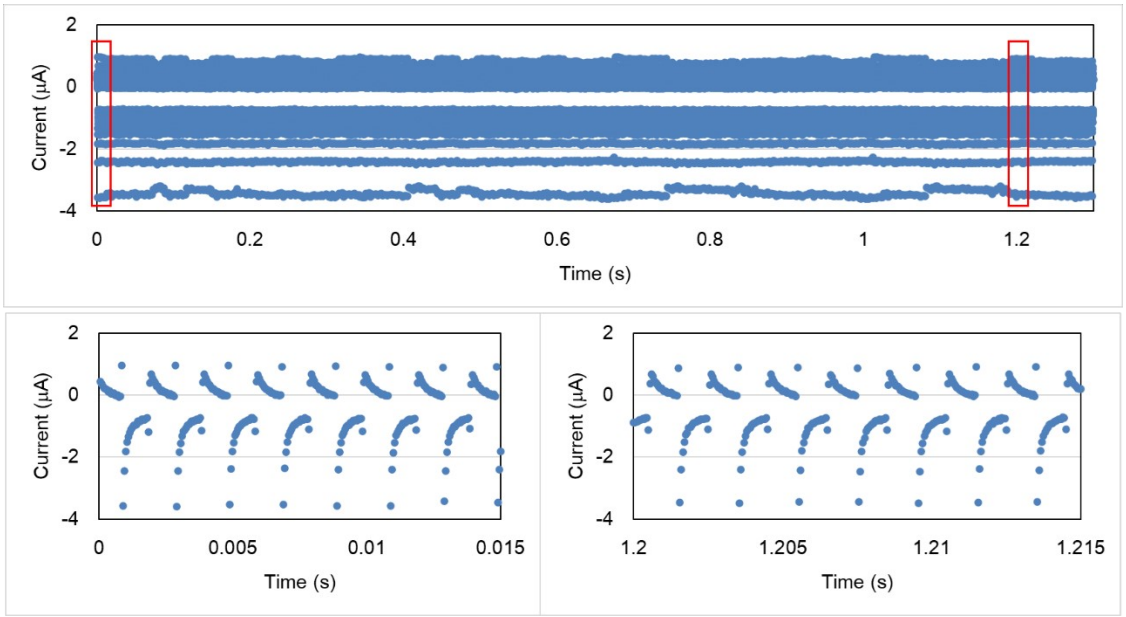

Figure S3. Time-dependent photoresponse of amorphous In_2S_3/NiO device under light illumination of 420 nm with pulsed frequency of 500 Hz. Red markers are highlighted in the bottom panel confirm stable operation over the prolonged operation.

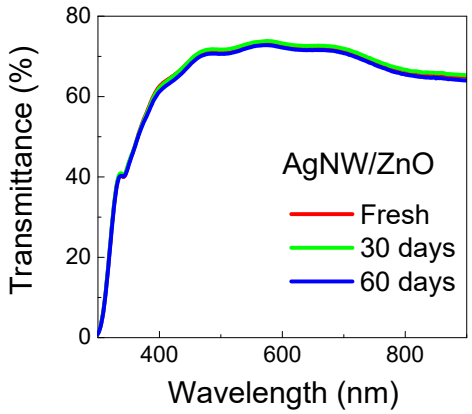

Figure S4. Transparency retention tests of AgNW/ZnO electrodes under ambient conditions.